Modelling Nonconductive Coating Based on EIS and WMR Tools

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Abstract

This paper represents the second part of a study aiming at developing a new tool to enhance the quality of nonconductive coatings on monitoring and modelling. The goal of this paper is to improve the modelling process of nonconductive coatings by integrating time-frequency domain features using wavelet Multi-resolution (WMR) based tool and the traditional Electrochemical Impendence Spectroscopy (EIS) measurements. Enamel coatings on aluminum substrates are excited with high frequency voltage source of a square wave using three-electrode arrangement. Signals of non-stationary features of the current response of enamel coated samples are analyzed using a windowing wavelet-based tool. The EIS data is utilized to estimate the initial values of the coating circuit model and the windowing wavelet transform is used to correlate laboratory and simulated data in order to define an accurate circuit model. The proposed technique is implemented on large sets of laboratory data of enamel coated samples, which shows excellent results.

Keywords

Nonconductive Coatings; Electrochemical Impedance Spectroscopy; Coating Modelling; Wavelet Multi-resolution Analysis; Fast Fourier Transform; Kaiser's Window

Introduction

Protective nonconductive coatings on metal substrates can be evaluated using various approaches including outdoor exposure, accelerated tests, sensitive measurement techniques for early detection, and combinations of accelerated tests and sensitive detection methods. Coating deterioration monitoring is further emphasized due to the rapid development of new coating formulations prompted by health and environmental regulations on hazardous coating constituents. The sought testing techniques should have attributes of reliability, sensitivity, and speed. The main objective of these tests from a manufacturing perspective is to reliably forecast the service life and performance characteristics of a coating in a short period of time. Traditional electrochemical impedance spectroscopy (EIS) is used widely to evaluate the resistance of coated metals to corrosion. A remarkable advantage of EIS over other laboratory techniques is the possibility of using very small amplitude signals without disturbing the properties measured and the possibility of working in low- or variable-conductivity environments.

Monitoring EIS spectrum can reveal any deterioration of the coating caused by exposure to an electrolyte and quantify the increase in corrosion rate of the underlying substrate due to this deterioration. These features of EIS can be used to model the processed sample and investigate the quality of the coating.

While EIS is used to extract features under steady-state source for sample modeling, there are important features which can be extracted during transient periods. These features disappear as fast as the excitation voltage/current jumps from a state to another and hence of a transient nature. The application of a square voltage source with high frequency generates voltage and current information that spread-over a wide-frequency band as the source reverse sits polarities. Such transient features reflect the response of the coated sample to the excitation source which can be integrated with the steady-state response extracted from EIS technique to generate an accurate model of the nonconductive coating.

The goal of this research is to develop a new tool to enhance the quality of nonconductive coatings on monitoring and modelling. The work is divided into two parts. The time-frequency domain features using wavelet-based tool and the traditional Electrochemical Impendence Spectroscopy (EIS) measurements are integrated to develop an enhanced test method that rapidly screens the nonconductive coating and gives measure on its quality. The objective of this paper is to develop an enhanced test method to model the

nonconductive coating. The proposed modeling process depends on comparing features extracted from fresh samples (reference) with others from deteriorated ones. The EIS data in the commissioning stage are utilized to define the initial values of the circuit model whereas fast Fourier transform (FFT), wavelet multi-resolution analysis (WMRA) and windowing wavelet multi-resolution (WWMRA) are used to tune these parameters toward an accurate circuit model as the nonconductive coating deteriorates.

Experimental Setup

Samples of 3003 aluminum alloy panels coated with porcelain enamels are prepared to define their electrical circuit model. The enamels used to produce the coating of the samples are of similar compositions but of different firing temperatures. Frit is the basic material of the enamel slip which is applied to aluminum alloy panels to produce the enamel coatings. Enamel slip is prepared by milling the frit with water, other ingredients and coloring oxides. The enamel composition of the first panel is fired at temperature of 500^{o}C (500^{o}C- sample) which is considered to have inferior quality. The enamel composition of the second panel is fired at temperature of 540^{o}C (540^{o}C- sample) to produce a coating of good characteristics.

EIS Measurements

During EIS laboratory investigation, a steady-state AC voltage with different frequencies is applied to various samples and the magnitude and phase of the impedance at each frequency, are used to monitor the quality of the coating. The EIS measurements are carried out at room temperature using a potentiostat/galvanostat Gill AC from ACM Instrument. A three-electrode arrangement consisting of a Ag/AgCl reference electrode, a platinum foil as a counter electrode and the coated sample as a working electrode are used. EIS measurements are performed by employing a sinusoidal potential perturbation of 30 mV at the open circuit potentials. The impedance spectra are measured with frequency sweep from 30.0 kHz to 0.001 Hz in logarithmic increment. The impedance diagrams are presented in the form of Nyquist and Bode plots. The extracted time-frequency domain information is then used to model the changes in the coating and the increase in the corrosion rate using different circuit models. A complete description of the EIS setup and measurement is published in part 1, Enhancing Monitoring of Nonconductive Coatings.

Wavelet-based Measurement Setup

Each of the two samples under investigation is placed under two Plexy-Glass tubes of 41.0mm diameter and 260.0 mm height and filled with 3% by weight sodium chloride solution at 20^{o}C. Both tubes are used initially to represent the reference sample of each coating (540^{o}C or 500^{o}C-fresh samples), one of which is cleaned with distilled water after taking the initial reference measurements, whereas, the second tube is frequently filled with the sodium chloride solution for long periods to impose deterioration on the underlying part of the sample (540^{o}C or 500^{o}C-old samples).

The voltage across the working electrode and the reference electrode is monitored using channel 1 of Tektronix oscilloscope TSD2120D, 100MHz through a voltage probe of 10Mhomresistance. The current through the working electrode is monitored using a current transformer LEM, type LA25-NP, 150kHz and its signal is captured using channel 2 of the oscilloscope. Agilent 33120A,100MHz function generator is used to feed the system with a rectangular excitation voltage waveform with controllable frequency and amplitude. A PCI Daq system including block terminal connector SCB-68 and NI-PCI 6220, multifunction M series, 250KS/S utilized to capture the function generator square signal is regarded as a reference.

The excitation voltage amplitude is varied between 5 to 1000 mV and its frequency is selected as 60 kHz, 30 kHz, and 15 kHz. These excitation frequencies, captured data size, and sampling rate are carefully selected to have the important features centered at certain resolution level (frequency bands) in the wavelet-based proposed technique.

Coating Circuit Model

To model the physical changes of the non-conducting coating material, suitable equivalent-circuit topologies have to be defined. Based on the underlying physical processes, the equivalent circuits should allow an optimum representation of the measured signals and the extracted features with a minimum set of model parameters. The coated sample is modeled as a collection of electrical elements and used to quantify the changes between the two samples (540oC or 500oC) as well as monitor the coating degradation within each sample (fresh and old samples). The simple circuit model shown in Fig.1 is utilized to simulate the coating failure.

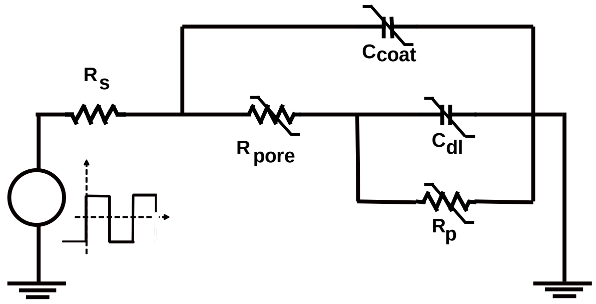

FIG.1 CIRCUIT MODEL OF COATED SAMPLE

The model includes five parameters. A resistance (R_s) is used to simulate the electrolyte resistance. The capacitance of the coating is represented using the coating capacitance (C_{Coat}). Any change in the resistance of coating due to penetration of electrolyte solution into the micropores of the coating is simulated using the pore resistance (R_{pore}). The corrosion rate of the metal substrate beneath the coating is represented by the polarization resistance (R_p). The metal-electrolyte interface due to the electrolyte charge and electrode charge is modeled by the double layer capacitance (C_{dl}). The capacitance of a typical undamaged coating with good barrier properties is about 1 nF/cm², however, for a coating that is under attack by an electrolyte, the values of the circuit elements will change (sometimes dramatically) as attack proceeds, which leads to changes in the sample response.

The Proposed Monitoring Technique

Wavelet multi-resolution analysis (WMRA) has been successfully applied in the analysis of transient features of many engineering systems. It is used to analyze electrochemical potential noises and reveal the characteristics of potential signals in detail, distinguish accurately the inhibiting effects of pigments and corrosion processes on the surface, and monitor the onset of localized corrosion. The wavelet transform (WT) offers an enhanced ability for discriminating among electrochemical noise signals arising from different types of localized corrosion.

The discrete wavelet transform (DWT) represents the signal $i(t)$ as a series of approximate $c_j(k)$ and detail $d_j(k)$ coefficients.

$$i(t) = \sum_k c_o(k)\phi\ (t-k) + \sum_k \sum_{j=0}^{J-1} d_j(k) 2^{j/2} \psi(2^j t - k) \quad (1)$$

If the scaling function and wavelets form an orthonormal basis, then according to Parseval's theorem, the energy of the signal can be partitioned in terms of the expansion coefficients. Fig. 2 shows the wavelet multi-resolution decomposition of the current through the working electrode. The signal is sampled at 10.0 MHz and decomposed into 8 resolution levels (D1 to D8) using db 40.

Significant improvement in the monitoring efficiency can be achieved by increasing the similarity of a single expansion system ($\chi_{k,n}(t)$) and the processed signal. A window function $w_k(t)$ is used in order to have a processed signal which resembles the selected mother wavelet and hence strengthening the important criteria in signal processing. The windowing version of the signal can be represented in terms of windowing version of the expansion coefficients $wc_j(k)$ in the form:

$$wf(t) = \sum_{k,n} wc_j(k)\chi_{k,n}(t) \quad (3)$$

The variable k is the time index while n is a frequency index. By requiring orthogonality of the basic functions, the expansion coefficients are computed by an inner product as:

$$wc_j(k) = \langle w_k(t) f(t), \psi(t) \rangle \quad (4)$$

Kaiser's window of length w_N is selected in the windowing process, which is mathematically presented as:

$$w[n] = \frac{I_o\{\beta[1-(n-u)^2/u^2]^{0.5}\}}{I_o(\beta(0,\alpha))} \quad for\ \ n = 0,1,....,w_N - 1 \quad (5)$$

where $I_o(x)$ is the modified Bessel function, u is the midpoint of the window function. The advantage of selected Kaiser's window is the ability to adjust the window shape by changing α parameter.

Using Mallat's algorithm, the set of detail expansion coefficients resulting from a windowing version of the signal at certain resolution can be defined as:

$$wd_j(k) = \sum_m h_1(m-2k)\ wc_{j+1}(m) \quad (6)$$

Then the dynamic behavior of the magnitude variation of the processed signal at different resolution levels is measured using the following relation:

$$Mag_j(w_c) = A \times Max\left|\sum_m h_1(m-2k)\, wc_{j+1}(m)\right| \quad (7)$$

Where the absolute value of the maximum coefficient localized at certain resolution level and at certain

center of Kaiser's window (w_c) is used to measure magnitude variation. The factor A is used to compensate for a constant energy as the scale changes to decompose the signal at higher or lower frequency resolutions.

The accuracy of the proposed monitoring tool is further enhanced by increasing the similarity between the processed signal and the selected mother wavelet. This is can be achieved by defining the best mother wavelet and optimal processing window size and shape as well as the sliding rate. The window size, w_N, is varied within a defined interval while fixing the other two parameters, window shape (α) and the sliding rate (N_s). For each set of windowing parameters, the pattern of the extracted signals is monitored. Choose w_N that produces the highest number of correct localization of correct pattern. Vary N_s and then α while keeping w_N fixed at the optimal value to enhance the extracted pattern.

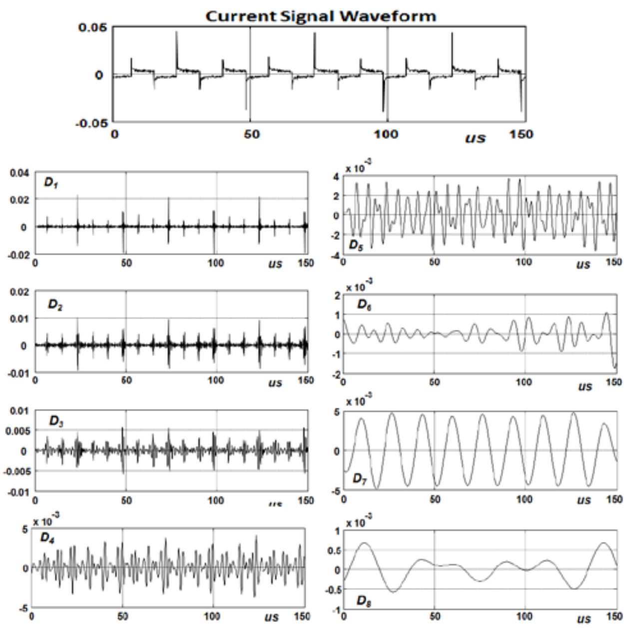

FIG. 2 WAVELET MULTI-RESOLUTION DECOMPOSITON OF WORKING ELECTRODE CURRENT

The Proposed Modeling Technique

Fig.3 shows the proposed procedure to enhance the accuracy of defining the coating circuit model. The accuracy of the simulated parameters (R_{pore}, R_p, C_{Core} and C_{dl}) is enhanced by minimizing the error in the current signals between the actual sample ($i(t)_{Lab}$) and the simulated one ($i(t)_{Simu}$) while modifying the circuit model parameters. The error is minimized while data is processed in frequency domain using FFT, the time-frequency domain using wavelet multi-resolution analysis (WMRA) and the maximum coefficients of windowing wavelet multi-resolution (WWMRA). The following steps summarize the proposed modeling technique:

1. The proposed technique has two inputs. Use the EIS data and PSCAD software simulator to construct the initial simulated circuit model and generate the simulated current signal $i(t)_{Simu}$. Use the proposed DSP based coating monitoring tool to generate the actual laboratory signal $i(t)_{Lab}$
2. Apply FFT to both simulated $i(t)_{Simu}$ and actual current laboratory signal $i(t)_{Lab}$ and minimize the error by modifying the simulated PSCAD circuit parameters.
3. Apply WMRA and compare the norm of the wavelet coefficients of i_{Simu} and i_{Lab} then minimize the error by adjustment of the simulated PSCAD parameters.
4. Apply WWMRA to the laboratory signal i_{Lab} assuming certain parameters ($\alpha, w_N, N_S \;\&\psi$). Define the optimal parameters that satisfy minimum errors $\Delta Mag_j \;\&\; \Delta Mag_R$.
5. Using the optimal parameters ($\alpha, w_N, N_S \;\&\psi$) define the estimated signals at the 7th resolution of the high energy for both the simulated and laboratory signals. Minimize the error by modifying the simulated PSCAD circuit parameters till getting maximum similarity between the two signals in terms of magnitude, shape and phase shift.

The optimal window parameters are defined and used to estimate the coating circuit model as shown in Fig. 1. The circuit model is simulated using PSCAD/EMTDC package, which is an electromagnetic time domain transient simulation environment and study tool. A random noise is added to the simulated data to imitate the field environment. The results are compared with the laboratory data generated under square wave source signal of 60 kHz, 30 kHz and 15 kHz. The initial values of the circuit model parameters (coating capacitance, pore resistance, polarization resistance and double layer capacitance) are approximated from the results of EIS technique. These parameters are adjusted till the FFT error, the $\| d_j(k) \|$ error and difference between the original laboratory signal and

the simulated signal extracted using proposed technique (WWMRA) as indicated in (7)is minimal.

In the windowing wavelet multi-resolution analysis (WWMRA) processing stage, the best mother wavelet and optimal windowing parameters are obtained by varying the window size, w_N, while fixing the other two parameters, α and N_s. For each set of windowing parameters, the pattern of the extracted signals as indicated in (7) is monitored by measuring the slope of the extracted signals (ΔMag_j) at the 7th and the 8th resolutions as well as their ratios.

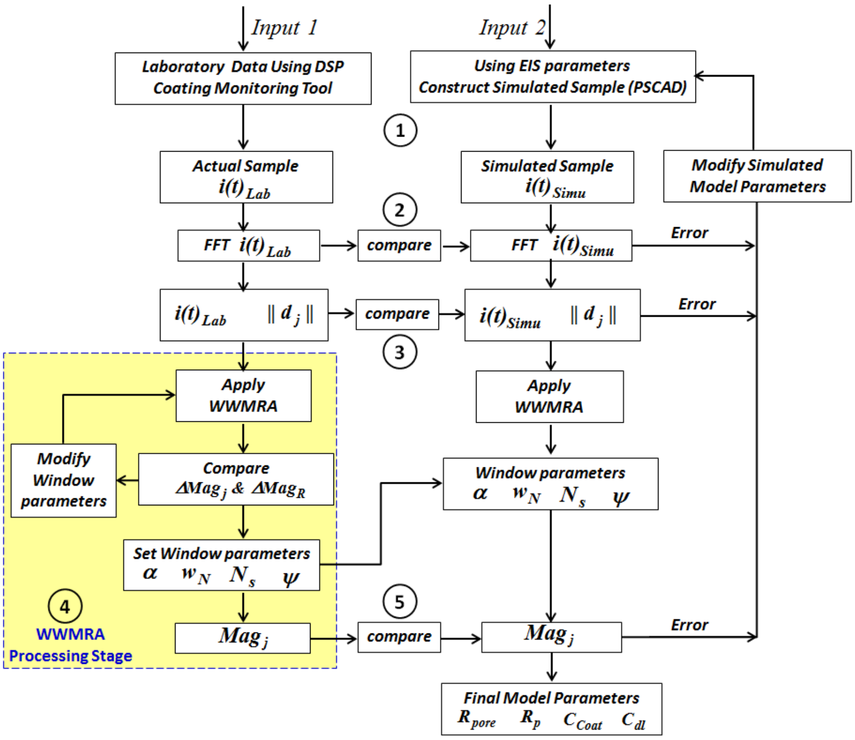

FIG. 3THE PROPOSED TECHNIQUE IN ESTIMATING COATING CIRCUIT PARAMETERS.

Application and Results

Fig.4 shows the results of implementing the proposed technique in defining the circuit model parameters of a fresh 500°C sample. The blue color represents the results of the laboratory signal and the red one represents the simulated signal. The waveforms of the laboratory and simulated working electrode currents, during 60kHz square wave source of 1.0 volt peak, are presented in Fig.4a. The FFT for a frequency band up to 1.0 MHz and the norm of wavelet coefficients for the 4th resolution level (312 to 625 kHz) up to the 10th resolution level (4.9 to 9.8 kHz) of simulated and laboratory data are presented in Figs.3b and c.

The main features of the processed signals are localized in the 7th resolution (39.1 to 78.1 kHz). Fig.4d shows the extracted signals using WWMRA, at the 7th resolution of the simulated and laboratory data. The circuit model parameters are adjusted till a minimum error between the processed laboratory data and the simulated model data is generated.

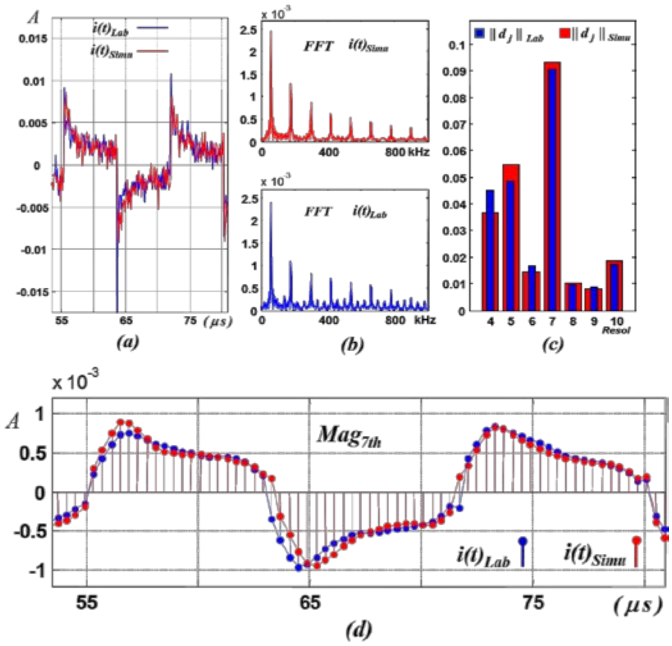

FIG.4 COMPARING THE SIMULATED AND LABORATORY DATA OF 500OC FRESH SAMPLE, A –SIGNALS WAVEFORM, B – SIGNALS FFT, C- SIGNALS WMRA NORM OF EXPANSION COEFFICIENTS AND D –WWMRA EXTRACTED SIGNALS AT 7TH RESOLUTION.

Implementation Issues and Discussion

The initial values of the double layer capacitance and the polarization resistance in circuit model are initiated using the Randles cell provided by ACM instrument.

The effect of varying the excitation voltage magnitude, frequency and waveform type is investigated. Fig.5shows the voltage (10.0 mV) across the counterelectrode and the reference electrode as well as the working electrode current during low excitation square voltage of 15.0 kHz. The monitored voltages and currents have DC components which are ignored during signal's processing. The voltage waveforms for fresh and old samples (Fig.5a and c) show different slopes that might be used in modeling the circuit elements. However, as the excitation voltage magnitude decreases, the working electrode currents of the fresh sample (Fig.5b) and old sample (Fig.5d) are corrupted with high noise levels that mask the main features of the current signals. The de-noising capability of the proposed technique can be utilized to overcome this problem. However, the maximum allowable source voltage with respect to the insulation coating type and thickness should be investigated. The effect of using sinusoidal, triangular waveforms or single excitation pulse should also be investigated.

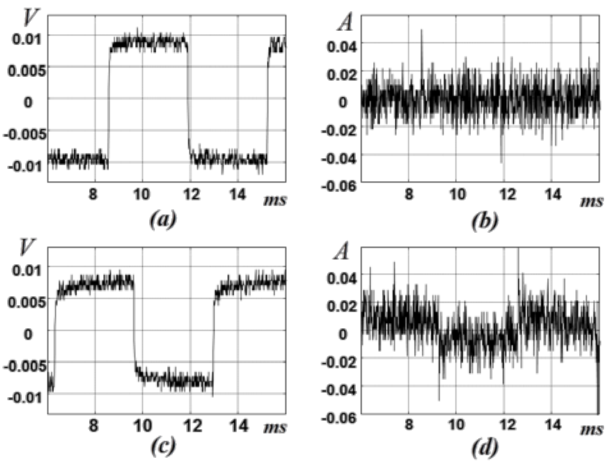

FIG.5 THE EFFECT OF LOW EXCITATION SOURCE MAGNITUDE ON OLD AND FRESHSAMPLES.

In the windowing wavelet multi-resolution analysis (WWMRA) processing stage, the best mother wavelet and optimal windowing parameters are obtained by varying the window size, w_N, while fixing the other two parameters, α and N_s. For each set of windowing parameters, the pattern of the extracted signals as indicated in (7) is monitored by measuring the slope of the extracted signals (ΔMag_j) at the 7th and the 8th resolutions as well as their ratios (ΔMag_R) which is mathematically presented as:

$$\Delta Mag_j(w_c) = Mag_j(w_c) - Mag_j(w_c - N_s) \tag{8}$$

$$\Delta Mag_R(w_c) = \frac{\Delta Mag_7(w_c)}{\Delta Mag_8(w_c)} \tag{9}$$

Choose w_N that produces the lowest ΔMag_j and ΔMag_R. Vary N_s and then α while keeping w_N fixed till the optimal mother wavelet and all other window parameters are defined. Fig.6 shows the results of ΔMag_7, ΔMag_8 and ΔMag_R for the following three mother wavelets and window parameters:

a- Rectangular window ($\alpha = 0$), $w_N = 120$, $N_s = 4$, *Coif 2* mother wavelet

b- Kaiser's window ($\alpha = 5$), $w_N = 85$, $N_s = 4$, *db4* mother wavelet

c- Kaiser's window ($\alpha = 85$), $w_N = 150$, $N_s = 4$, *db10* mother wavelet

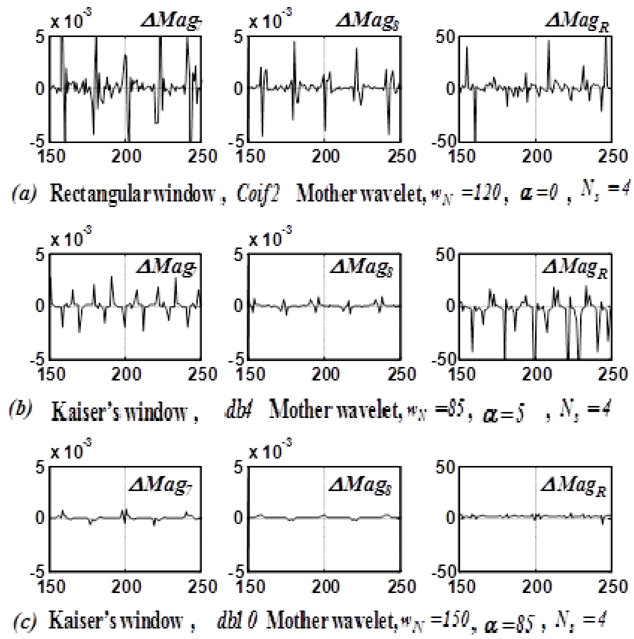

FIG.6 THE VIBRATION IN THE SLOPES ΔMag_7 AND ΔMag_8 AND THEIR RATIO ΔMag_R FOR DIFFERENT WINDOWING PARAMETERS.

The results of simulated circuit parameter adjustment in order to minimize the error ΔMag_7, ΔMag_8 and ΔMag_R are shown in Fig. 7. These parameters are adjusted by trial and error till the simulated and laboratory data match. The mathematical relation that reflects the simultaneous changes in the circuit parameters, as the coating degrades, should be investigated.

The proposed technique is also implemented to model the same sample after exposing it to sodium chloride solution for a long period. The same window parameters are used and the circuit elements that model the organic coating are adjusted till minimum error condition is obtained. The resulted circuit model parameters of the proposed technique of the fresh 500^{o}C and the old 500^{o}C samples are presented in Table 1.

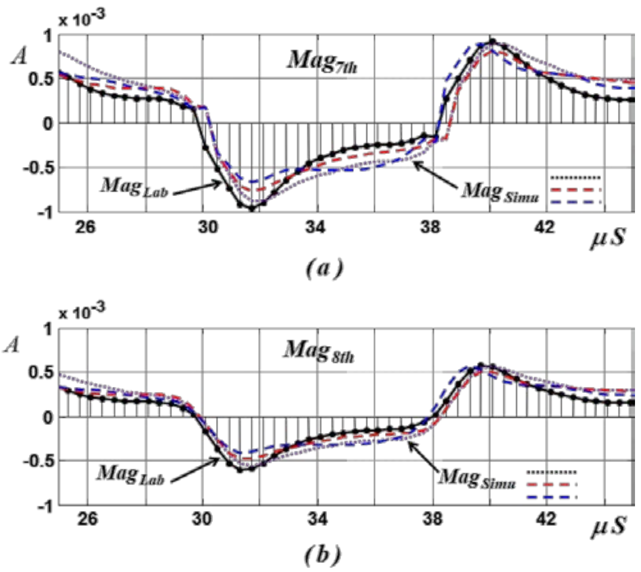

FIG.7 THE EFFECT OF VARYING SIMULATED CIRCUIT PARAMETERS ON THE EXTRACTED CURRENTS AT (A) THE 7TH AND (B) THE 8TH RESOLUTIONS

TABLE 1ESTIMATED PARAMETERS OF THE 500^{O}C FRESH AND OLD SAMPLES

Sample Type	R_p $(k\Omega)$	R_{pore} $(k\Omega)$	C_{Coat} (μF)	C_{dl} (μF)
500^{o}C Fresh	38.60	21.00	0.80×10^{-2}	92.20
500^{o}C Old	20.00	14.665	5.00×10^{-2}	100.00

Conclusion

This paper proposes an enhanced tool to model non-conducting coatings. A wavelet-based tool is integrated with the conventional EIS in order to enhance the circuit model. The EIS data will be implemented on specific locations to define the initial values of the circuit model. The initial simulated circuit model is excited with a square wave and its working electrode current response is compared with the laboratory results. The circuit model parameters are adjusted until a minimum error in the frequency domain is achieved using FFT and wavelet-domain by WMRA. The non-stationary nature of extracted laboratory data and simulated data are also compared using WWMRA. The proposed technique is implemented on large sets of laboratory data, which shows excellent results.

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